LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

LLNL-CONF-674722

Direct Detection of Pu-242 with a Metallic Magnetic Calorimeter Gamma-Ray Detector

C. Bates, C. Pies, D. Hengstler, A. Fleischmann, L. Gastaldo, C. Enss, S. Friedrich

July 16, 2015

16th International Workshop on Low Temperature Detectors
Grenoble, France
July 20, 2015 through July 24, 2015

Disclaimer

Direct Detection of Pu-242 with a Metallic Magnetic Calorimeter Gamma-Ray Detector

C. Bates[1]* • C. Pies[2] • D. Hengstler[2] • A. Fleischmann[2] • L. Gastaldo[2] • C. Enss[2] • S. Friedrich[1]**

[1] Lawrence Livermore National Laboratory, 7000 East Ave., L-188, Livermore, CA 94550, USA
[2] Universität Heidelberg, Kirchhoff Institut für Physik, INF 227, 69120 Heidelberg, Germany

Abstract Cryogenic high-resolution γ-ray detectors can improve the accuracy of non-destructive assay (NDA) of nuclear materials is cases where conventional high-purity germanium detectors are limited by line overlap or the Compton background. We have improved the performance of gamma detectors based on metallic magnetic calorimeters (MMCs) by separating the $0.5 \times 2 \times 0.25$ mm^3 Au absorber from the Au:Er sensor with sixteen 30 μm diameter Au posts. This ensures that the entire γ-ray energy thermalizes in the absorber before heating the Au:Er sensor, and improves the energy resolution at 35 mK to as low as 90 eV FWHM. This energy resolution enables the direct detection of γ-rays from Pu-242, an isotope that cannot be measured by traditional NDA and whose concentration is therefore inferred through correlations with other Pu isotopes. The Pu-242 concentration of 11.11 ± 0.42 % measured by NDA with MMCs agrees with mass spectrometry results and exceeds the accuracy of correlation measurements.

Keywords Metallic magnetic calorimeters • Nuclear safeguards • Gamma spectroscopy • Non-destructive assay • Plutonium isotopics • Pu-242

1. Introduction

Safeguarding nuclear materials requires the accurate assay of fissile isotopes throughout the nuclear fuel cycle [1]. Destructive assay (DA) based on mass spectrometry provides the highest accuracy, but requires the time consuming chemical separation of different elements. Non-destructive assay (NDA) based on γ-spectroscopy with high-purity germanium (HPGe) detectors is less accurate, but it can be automated and is faster and less expensive. NDA

* Current address: Los Alamos National Laboratory, NM 87545, USA
** Corresponding author: friedrich1@llnl.gov

is therefore preferred as long as it can provide the required accuracy for a specific application. One limitation of NDA with HPGe detectors is that it cannot detect γ-signatures of Pu-242, because the Pu-242 γ-rays are weak and lie in close proximity to other much stronger Pu lines. The concentration of Pu-242 is therefore typically inferred indirectly through correlations with the concentration of other Pu isotopes [1, 2]. However, these correlations introduce systematic errors, especially for high burn-up fuel with Pu-242 concentrations between 5 and 10% [2]. These errors are unacceptably high when large amounts of Pu need to be assayed, like at reprocessing plants that process tons of Pu from nuclear fuel each year.

Cryogenic γ-detectors can overcome some of the limitations of HPGe detectors in nuclear safeguards due to their higher energy resolution and higher peak-to-background ratio [3-7]. This motivates our development of γ-detectors based on metallic magnetic calorimeters [7]. Here we illustrate the use of MMC gamma detectors in a nuclear safeguards application with a direct measurement of Pu-242 γ-rays, and show that our MMCs have sufficient energy resolution for NDA of Pu samples that include Pu-242.

2. Experiment

The MMC γ-detector fabrication is based on the maXs-200 detector design developed at the University of Heidelberg [5]. It uses two 2 mm × 0.5 mm Au:Er sensors with 600 ppm of ^{166}Er dopants in a gradiometer geometry. The sensors are deposited on top of a Nb meander coil that is used to apply the magnetic field to split the Zeeman levels of the Er dopants and to pick-up the gamma-induced change in magnetization. To adapt these devices for high-resolution γ-detection, we have developed a process that separates the 250-μm-thick Au absorber from the Au:Er sensor by 16 Au posts with a diameter of 30 μm. This bottleneck ensures that the entire γ-energy is thermalized in the absorber before heating the Au:Er sensor, and prevents line broadening due to the escape of athermal phonons into the Si substrate.

The post dimensions are defined using AZ 5214E negative photoresist, and the mold for the absorber is formed with a spin-casting process using AZ 125 nXT photoresist [8]. The resist is spun on at 1500 rpm for 2 s for a mold thickness of 300 μm, baked for 6 h at a temperature of 105 °C, exposed to 365 nm UV light at 10 J/cm^2, and developed using AZ 326 MIF. After ion-cleaning the underlying Au:Er surface to ensure good thermal coupling, the Au posts and absorber are electroplated onto the Au:Er sensor at a rate of ~0.3 μm/ min to a thickness of 250 μm.

To demonstrate the performance of these detectors in nuclear safeguards applications, we prepared a mixed-isotope Pu source by adding Pu-242 to a

solution of weapons-grade plutonium that consisted of Pu-239, Pu-240 and small quantities of impurities and decay products. Destructive assay by mass spectrometry showed 83.75 ± 0.13 % Pu-239, 5.39 ± 0.01 % Pu-240 and 10.81 ± 0.02 % Pu-242. The solution was dried inside a small Al holder that could be closed with a thin Al lid for containment. The source was mounted in the adiabatic demagnetization refrigerator (ADR) outside a small hole in the Nb shield that contained the MMC and the first-stage SQUID preamplifier, so that its decay produced γ-rays at a rate of a few counts per second in the MMC. Since the energy resolution of the MMC degrades with increasing temperature, the ADR was not temperature regulated, but operated at its base temperature of ~35 mK and allowed to warm by a few mK during data acquisition. This allowed ~8 to 10 hours of data acquisition per ADR cycle, but required a drift correction to compensate for the reduced pulse height at higher temperature. For this, a running average of 80 events was calculated for one of the strong intermediate-energy lines in the spectrum, such as the Am-241-line at 60 keV, and all other lines were scaled by this average. The linearity of the MMC response allowed applying the same correction to minimize the width of all lines in the spectrum.

All γ-induced waveforms of the two pixels were captured, separated according to their polarity and processed off-line with a trapezoidal filter. The energy was calibrated with a linear fit to the Gaussian centroids of the strongest lines in the spectrum, and 24 spectra from 12 ADR cycles could be added to improve the statistical accuracy of the measurement without loss in the energy resolution of 130 eV FWHM (Fig. 1).

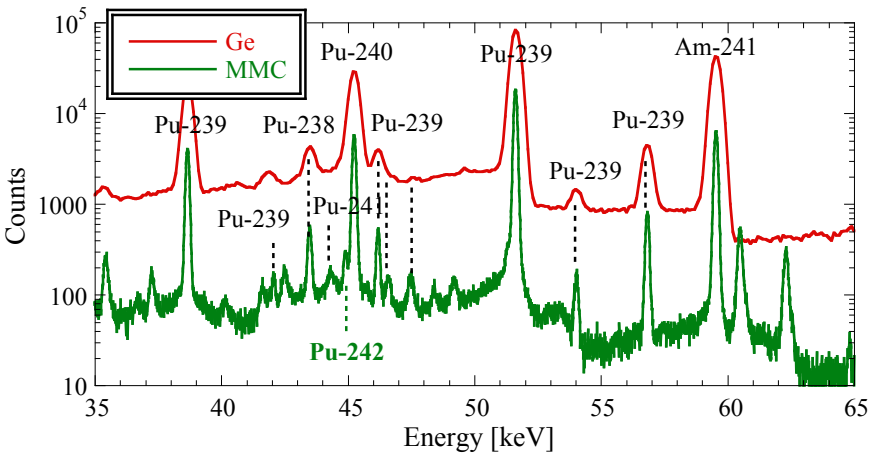


Fig. 1 MMC γ-spectrum of the Pu source (*bottom*), compared to that of a statistics-limited Ge detector (*top*). Unlabeled lines are due to the escape of Au and Ge $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays from the two detectors. (Color figure online.)

It contains ~1.5×10^6 events, and is dominated by emissions form the isotopes of Pu-239 and Pu-240 as expected. The small signal at 44.915 keV is the only signature in the spectrum that indicates the presence of Pu-242. It is hidden in the wings of the much stronger Pu-240 emission at 45.244 keV when using a Ge detector, but clearly separated in the MMC spectrum (Fig. 1 inset). This allows using the MMC spectrum for quantitative analysis of the Pu source.

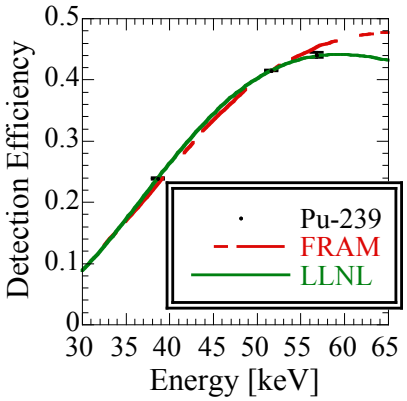
3. NDA with MMCs

Non-destructive assay (NDA) of nuclear materials determines the abundance of isotopes from the measured intensity of the corresponding γ-rays, taking into account the branching ratios of the different lines and their relative detection efficiency. The detection efficiency changes as a function of energy depending on the composition and the dimensions of the source, the detector and the intervening materials. Uncertainties in relative detection efficiency therefor cause systematic errors in the assay, especially if the composition of the source and thus its absorption properties are unknown.

In addition, current NDA codes such as FRAM always assume the use of a Ge-based γ-detector, whose detection efficiency is substantially different from that of a Au-based MMC detectors. This causes assays based on existing NDA codes to provide isotopic compositions that systematically differ from the DA (mass spectrometry) values by several standard deviations. We have therefore used the measured intensities of the strong Pu γ-rays at 38.666, 51.624 and 56.836 keV (cf. Fig.1) and the known branching ratios of these lines to calculate a custom curve of the detection efficiency. For this relatively small energy range we can model the detection efficiency as a product of self-absorption in the Pu source (thickness d_{Pu}), transmission of the Al source holder (d_{Al}) and absorption of the Au absorber (d_{Au}) according to

$$\text{Efficiency} = \text{const} \cdot \frac{1 - e^{-\mu_{Pu} d_{Pu}}}{\mu_{Pu} d_{Pu}} \cdot e^{-\mu_{Al} d_{Al}} \cdot (1 - e^{-\mu_{Au} d_{Au}}) \qquad (1)$$

The data used for the energy-dependent absorption coefficients μ were taken from the ENDF VII.1 photo-atomic library [9], using the total photon cross section (MT 501) to calculate absorption in the Pu source and the Al holder, and the photoelectric cross section (MT 522) to calculate the efficiency of the Au absorber. We then use a least square fit to extract the effective thickness d_{Pu} of the Pu source and the proportionality constant in Eq. (1). The resulting efficiency differs slightly from the Ge-based curve in FRAM by several percent in the energy range of interest (Fig. 2).



Pu Isotope Analysis

Isotope	DA [%] (Mass Spec)	NDA [%] (MMCs)
Pu-239	83.75 ± 0.13	83.67 ± 0.42
Pu-240	5.39 ± 0.01	5.22 ± 0.11
Pu-242	10.81 ± 0.02	11.11 ± 0.42

Fig. 2 (left): Detection efficiency for the experimental setup according to Eq. 1 (*solid line*) and according to FRAM (*dashed line*). **Table 1** (right): Isotope concentrations of the Pu sample as determined by non-destructive assay (NDA) with MMCs, compared to the values from mass spectrometry (DA).

Based on this efficiency curve, the Pu concentrations of the sample from NDA with MMCs agree with the reference values obtained by DA within the uncertainty of the measurement (Table 1). This includes the concentration of Pu-242 that cannot be detected directly with Ge detectors and whose relative accuracy of 0.42/11.11 = 3.8 %is already better than typical correlation measurements [2]. At this point, the Pu-242 uncertainty is still limited by the statistical error in the number of counts in the Pu-242 peak, so that it can be improved by with detector arrays.

In summary, we have built an MMC γ-detector whose Au absorber is separated from the Au:Er sensor by sixteen 30 μm diameter Au posts so that the γ-energy fully thermalizes in the absorber. Its energy resolution of 130 eV FWHM at 35 mK is sufficient to directly detect the weak Pu-242 γ-ray at 44.915 keV and enable NDA on Pu samples that contain Pu-242. The accuracy of ±3.8% is currently still limited by the counting statistics of the Pu-242 line, and can be improved by developing MMC γ-detector arrays.

Acknowledgements We thank R. Henderson for fabricating the mixed-isotope Pu source. We gratefully acknowledge funding by the U.S. Department of Energy Office of Non-Proliferation Research NA-22 under grant LL12-MagMicro-PD2Ja. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, *Passive Non-Destructive Assay of Nuclear Materials*, Office of Nuclear Regulatory Research (NUREG/CR-5550), Washington, 1991
2. G. Bignan, W. Ruhter, H. Ottmar, A. Schubert, C. Zimmerman, *ESARDA Bulletin* **28**, 16 (1998)
3. M. F. Cunningham et al., *Appl. Phys. Lett.* **81**, 159–161 (2002); DOI: 10.1063/1.1489486
4. W. B. Doriese et al., *Appl. Phys. Lett.* **90**, 193508 (2007); DOI: 10.1063/1.2738371
5. C. Pies et al., *J. Low Temp. Phys.* **167**, 269–279 (2012); DOI: 10.1007/s10909-012-0557-z
6. D. A. Bennett et al., *Rev. Sci. Inst.* 83, 093113 (2012); DOI: 10.1063/1.4754630
7. C. R. Bates, C. Pies, S. Kempf, L. Gastaldo, A. Fleischmann, C. Enss, S. Friedrich, *J. Low. Temp. Phys.* **176**, 631–636 (2014); DOI: 10.1007/s10909-013-1063-7
8. M. Staab, F. Greiner, M. Schlosser, H. Schlaak, *J. Microelectromech. Syst.* **20**, 794–796 (2011); DOI: 10.1109/JMEMS.2011.2159098
9. M. Chadwick et al., *Nucl. Data Sheets* **112**, 2887–2996 (2011), Special Issue on ENDF/B-VII.1 Library; DOI: 10.1016/j.nds.2011.11.002